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*Published in:*

Radiation Measurements

*DOI:*

[10.1016/j.radmeas.2015.01.013](https://doi.org/10.1016/j.radmeas.2015.01.013)

*Publication date:*

2015

*Citation for published version (APA):*

Timar-Gabor, A., Constantin, D., Buylaert, J. P., Jain, M., Murray, A. S., & Wintle, A. G. (2015). Fundamental investigations of natural and laboratory generated SAR dose response curves for quartz OSL in the high dose range. *Radiation Measurements*, 81, 150-156. <https://doi.org/10.1016/j.radmeas.2015.01.013>

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## \*Highlights (for review)

- Double saturation exponential laboratory dose response
- Behaviour is inherent to quartz
- Natural dose response curves enter saturation earlier than laboratory dose response curves
- Initial dose dependent sensitivity changes may lead to poor dose recovery at very high doses

# Fundamental investigations of natural and laboratory generated SAR dose response curves for quartz OSL in the high dose range

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## Abstract

SAR-OSL investigations on quartz from Romanian loess resulted in non concordant fine and coarse-grain ages for equivalent doses higher than ~100 Gy. The laboratory dose response for both grain sizes is well represented by a sum of two saturating exponential functions, fine and coarse grains characterised by  $D_{01}$  and  $D_{02}$  values of ~140 and ~1400 Gy and ~65 and ~650 Gy respectively. Pulsed OSL experiments confirmed that this behaviour is almost certainly inherent to quartz and not caused by contamination with another mineral. Natural dose-response curves do not follow the same pattern and enter saturation much earlier. Analysis of time resolved spectra indicated similar luminescence lifetimes for both fine and coarse quartz grains, and natural and laboratory generated OSL signals seem to use the same non-dose-dependent recombination pathways. The natural signals of a sample with an expected equivalent dose of 2000-2500 Gy were found to be below the saturation level of the laboratory dose response curve for both grain sizes; this also applied to the luminescence signals measured after >5000 Gy given on top of natural doses.

**Keywords:** quartz; CW-OSL; TR-OSL; SAR protocol; dose response.

## 1. Introduction

Many thousands of optically stimulated luminescence (OSL) ages have been produced in the last decade using quartz, mainly using the simple single-aliquot regeneration (SAR) protocol (Murray and Wintle, 2000) and the modified SAR protocol (Murray and Wintle, 2003). The basic and fundamental assumption of luminescence dating is that the growth of the luminescence signal in nature can be reproduced under laboratory conditions by performing irradiations with a calibrated beta or gamma source. This comparison is based on the assumption that trapped charge distribution is not dependent on the dose rate, type of radiation or irradiation history before deposition (i.e. before sunlight exposure of the sediment).

In the SAR procedure, a dose response curve is obtained by plotting the ratio of the OSL signal to the response to a test dose. If the OSL signal is derived from a single electron trap and recombination centre, then the form of the dose response curve would be a single saturating exponential (e.g. Bøtter-Jensen et al., 2003). Some such dose response curves have been reported (e.g. Wintle and Murray, 2006); however, many SAR dose response curves for OSL signals from fine grained quartz dominated by the fast component display continuing growth at high doses (e.g. Lowick et al., 2010, 2011; Kreutzer et al., 2012). This behaviour would, in theory, enable higher equivalent doses to be obtained. In practice however, few ages have been obtained by SAR-OSL dating that have been in agreement with independent age control beyond the Eemian. Using multiple grain SAR-OSL procedures, Buylaert et al. (2007) reported natural signals below the laboratory saturation level for a sample with an expected age of ~900 ka. To our knowledge, for quartz that displays a continuing growth at high doses, there are no reports in the literature of natural signals found in saturation.

Our previous studies using fine (4-11  $\mu\text{m}$ ) and coarse (63-90  $\mu\text{m}$ ) quartz extracted from Romanian loess have shown that natural OSL signals from infinitely old samples are significantly below the laboratory saturation level (Timar-Gabor et al., 2012). Also, it has been reported that ages derived from these two grain size fractions are mutually inconsistent for equivalent doses higher than about 100 Gy, in the case of quartz extracted from both the Lower Danube Basin and the Carpathian Basin (Timar-Gabor et al., 2011, Constantin et al., 2014, Constantin et al. in press, Timar-Gabor et al., in press). Moreover, our previous results derived from quartz extracted from Romanian loess indicate that the laboratory dose response curve does not accurately reflect the growth of the OSL signal in nature (Timar-Gabor and Wintle, 2013). The present work is directed at obtaining more insight into these observations, by performing additional continuous-wave OSL experiments, and by applying time resolved luminescence on quartz extracts; these experiments are designed (i) to test whether the observed luminescence growth is a characteristic of quartz rather than an artefact caused by feldspar contamination, and (ii) to determine whether luminescent recombination occurs at different centres at large doses.

## 2. Samples, instrumentation and measurement protocol

Experiments presented in the current study have been conducted on archived quartz separates (63-90  $\mu\text{m}$  and 4-11  $\mu\text{m}$  grains) from our previous investigations on Romanian loess. Samples coded as MV have been collected from Mircea Vodă section in Dobrudjea (Timar et al., 2010; Timar-Gabor et al., 2011). MST is the lab code attributed to Mostistea section from the Romanian Plain (Vasiliniuc et al., 2011; Timar-Gabor et al., 2012). CST samples were taken from Costineşti section near the Black Sea shore (Timar-Gabor and Wintle, 2013; Constantin et al., 2014), while samples coded as LCA come from Lunca section in South Western Romania (Constantin et al., in press). More details on these samples are given in the supplementary material (**Table S1**). Continuous-wave optically stimulated luminescence (CW-OSL) measurements on quartz were made in the Cluj Luminescence Dating Laboratory with a Risø TL/OSL DA-20 reader equipped with blue light emitting diodes

(LEDs) emitting at  $470 \pm 30$  nm and IR LEDs emitting at  $875 \pm 80$  nm; luminescence signals were observed through a 7.5 mm thick Hoya U-340 UV filter. Irradiations have been carried out using a  $^{90}\text{Sr}$ - $^{90}\text{Y}$  beta source, calibrated using fine and coarse quartz (e.g. Hansen et al., this issue). The dose rate (at the time of measurement) for coarse grains mounted on stainless steel discs was  $0.147 \text{ Gy}\cdot\text{s}^{-1}$  and  $0.118 \text{ Gy}\cdot\text{s}^{-1}$  for fine grains deposited on aluminium discs. Details of the TL/ OSL reader can be found in Thomsen et al. (2006). The OSL signal was collected in time intervals of 0.154 s. All samples have been analysed using a SAR protocol (Murray and Wintle, 2000, 2003). The OSL signal used for analysis was that obtained for the first 0.308 s of the decay curve minus a background derived from the signal measured between 2.464 and 3.080 s, as had been used in previous studies on Romanian loess (Timar et al., 2010; Timar-Gabor et al., 2011; Timar-Gabor et al., 2012; Timar-Gabor and Wintle, 2013) following the recommendations of Cunningham and Wallinga (2010). All OSL signals analysed were characterised by a rapid decay and the shape of the decay curve is not dose dependent (**Figure S1**). Our previous studies showed that the shape of the linearly modulated OSL signal is not dependent on the age of the sample (Timar-Gabor et al., 2011- figure 4), nor on either the administered dose (Timar-Gabor et al., 2012- figure S1 and S4) or the thermal treatment previously applied (Timar-Gabor et al., 2011- figure 5). This provides evidence that the behaviour reported is characteristic of quartz OSL signals dominated by the fast component, as also reported by other studies (Lowick et al., 2010). Natural ( $L_n$ ) and regenerated ( $L_x$ ) signals were measured after a preheat of 10 s at  $220^\circ\text{C}$ ; the corresponding responses ( $T_n$  and  $T_x$ ) to a test dose (16 Gy) were measured after a cutheat to  $180^\circ\text{C}$  unless otherwise stated. The value of the test dose was kept constant through all measurements for both grain sizes of quartz. A high-temperature bleach was performed after each SAR cycle by stimulating with the blue diodes for 40 s at  $280^\circ\text{C}$  (Murray and Wintle, 2003).

Time resolved optically stimulated luminescence (TR-OSL) experiments were carried out at DTU Nutech on a Risø TL/OSL-20 (designated as Reader V) equipped with an integrated pulsing option to control the LEDs, and a Photon Timer attachment to record the TR-OSL (Lapp et al., 2009). All TR-OSL experiments have been carried out using the same measurement parameters as used in the CW-OSL measurements. For stimulating and recording the TR-OSL signals, the total measurement time was set to 100 s, with a pulse period of 500  $\mu\text{s}$  consisting of an “on time” of 50  $\mu\text{s}$  and an “off time” of 450  $\mu\text{s}$ ; this is equivalent to a net stimulation period of 10 s in CW-OSL. A total of 500 data points was used for data collection in pulsed stimulation; the sample was not illuminated during the first and the last five channels. Measurement of dose response curves using pulsed OSL measurements was designed to reproduce the CW-OSL measurement conditions as closely as possible, and to minimise any possible feldspar contamination of the quartz, by: (i) selection of an on time of 50  $\mu\text{s}$  (ii) gating the photomultiplier for counting only during the off period; (iii) ignoring the first 4  $\mu\text{s}$  during the off time in TR-OSL OSL data analysis (Ankjærgaard et al., 2010).

### 3. Laboratory dose response of quartz OSL

#### 3.1. CW-OSL investigations

1 Previous studies on Romanian loess showed quartz saturation at large doses, especially in the case of fine grains.  
2  
3 Here we compare the laboratory dose response of fine and coarse grains up to 5kGy. For fine grains, 15 aliquots  
4  
5 (from different sections in Romania) are shown (**Figure 1a**) one aliquot of MV10 and CAC 2.1.; two aliquots of  
6  
7  $CST_{inf}$ , MST 2, MST 6, MST 13, MST 19 and three aliquots of LCA15; it can be seen that the saturation  
8  
9 characteristics are similar and all datasets can be well represented by a sum of two saturating exponential  
10 functions:

$$11 \quad I(D) = I_0 + A(1 - \exp(-D/D_{01})) + B(1 - \exp(-D/D_{02}))$$

12  
13  
14 Any scatter in the data may be due to different origins of the loess as samples come from both SE and SW  
15  
16 Romania. For coarse grains, 11 aliquots are shown (**Figure 1b**), one aliquot from samples CAC. 2.1,  $CST_{inf}$ ,  
17  
18 LCA 15 and two aliquots of samples MV8, MST3, MST 17, and MST 19. The scatter is greater than for the 16  
19  
20 samples taken down the section at Costineşti (Timar-Gabor and Wintle, 2013). This may be due to the higher  
21  
22 dose range investigated (5000 Gy in the present study compared to 1000 Gy in the previous work) but may also  
23  
24 suggest a continuity of source material at one site; in that case, **Figs. 1a** and **1b** may show a variation in source  
25  
26 material across southern Romania. Nevertheless, and despite the observed scatter, the saturation characteristics  
27  
28 of all samples from a specific grain size are rather similar, but the average behaviour of the two grain sizes is  
29 markedly different.

30  
31 The reproducibility of the dose response curve was also examined (**Figure S2**). For coarse grains from sample  
32  
33 MV8, the dose response curve was taken up to 15 kGy. After constructing this first dose response, the sample  
34  
35 was bleached, given a dose of 170 Gy and then the process repeated; the 2<sup>nd</sup> dose response agreed with the first.  
36  
37 As expected, the sensitivity ( $T_x/T_{nat}$ ) fluctuates, although the response to the constant test dose does not change  
38 by more than about 30% from the initial value.

39  
40 Saturation characteristics of the dose response curves are not influenced by the thermal treatment used for either  
41  
42 fine or coarse grains (**Figure S3**). The  $D_0$  values for different thermal treatments are given in **Table S2**. The  
43  
44 scatter in  $D_0$  is probably affected by the restricted number of points used to construct the dose response and the  
45  
46 maximum dose given; the latter is not high enough for the dose response to be fully saturated. Nevertheless, it is  
47  
48 clear that the saturation characteristics of the fine and coarse grains remain significantly different when  
49  
50 compared to each other no matter what thermal treatment is used. Contrary to the findings of Lai et al. (2008),  
51  
52 heating the samples to 400°C produced only a trivial change in the shape of the laboratory dose response curve  
53 (**Figure S4**).

54  
55 If fine grains are derived from the comminution of coarser grains, it is very likely that fine grains have  
56  
57 experienced many more cycles of bleaching and irradiation than coarse grains. This process is known to sensitise  
58  
59 coarse grains (Moska and Murray, 2006) and may change the shape of the dose response curve. To test this, nine  
60  
61 aliquots of coarse grains from different samples were given multiple cycles of bleaching (100 s blue diodes at  
62  
63 room temperature/ pause 10000s/100 s blue diodes at room temperature) and irradiation (beta dose). All aliquots  
64  
65

1 sensitized by 60% after 8 cycles of bleaching and irradiation (total dose of approximately 1000 Gy). However,  
2  
3 there was no noticeable change in the dose response curve shape constructed using the aliquots that were  
4  
5 subjected to this treatment compared to aliquots from the same sample on which the dose response curve had  
6  
7 been constructed following the measurement of the natural, at least up to doses of 1000 Gy (**Figure S5**).

8 A fundamental difference that exists in nature between coarse and the fine grains is the past irradiation history,  
9  
10 fine grains experience a significant alpha dose rate, in contrast to coarse grains. However, after repeated cycles  
11  
12 of alpha irradiation with doses up 120 kGy (4.8 kGy effective alpha dose, assuming an “a value” of 0.04) prior to  
13  
14 beta dose response construction, we were unable to detect any significant effect on the saturation characteristics  
15  
16 of previously-etched quartz grains; thus, it is unlikely that alpha irradiation is causing surface defects that might  
17  
18 have been responsible for the higher saturation characteristics of fine grains (Constantin et al., this issue).

### 21 3.2. TR-OSL investigations

22  
23 In a previous study (Timar-Gabor et al., in press QI), time-resolved OSL (TR-OSL) measurements were made  
24  
25 on fine and coarse grains from one sample of Serbian loess from Orlovat. The shape of the TR-OSL was typical  
26  
27 of quartz (Chithambo, 2003; Denby et al., 2006; Thomsen et al., 2008). In addition, the similarity of the shapes  
28  
29 with and without prior exposure to IR further confirmed that the OSL was derived from quartz and that no part  
30  
31 of the signal was from feldspar. Dose response curves for both grain sizes were obtained using the TR-OSL  
32  
33 signal and were found to be very similar to those using the initial part of the CW-OSL signal. The equivalent  
34  
35 doses were also in agreement (TR-OSL  $171 \pm 9$  and  $170 \pm 16$  Gy for fine and coarse grains, respectively; the  
36  
37 equivalent values for CW-OSL were  $175 \pm 4$  and  $181 \pm 10$  Gy). This study was repeated on fine and coarse grains  
38  
39 from four samples of loess of different ages from Costinești and Lunca sections. The samples were preheated to  
40  
41 220 °C and stimulated at 125 °C. The IR treatments consisted of stimulation for 200 s at 125°C. TR-OSL curves  
42  
43 for fine and coarse grains given different doses, recorded both during “on and off times” are presented in  
44  
45 **Figures 2a and 2b**, respectively. During the “off time”, the signals display a slow decay that can be fitted with  
46  
47 a single exponential function.

48  
49 The aliquots have been measured using a typical SAR protocol measurement sequence, with a test dose of 19  
50  
51 Gy, and the counts from the photomultiplier were gated to record only during the off period. Data were  
52  
53 registered in 500 channels of 0.2 seconds, each channel consisting of 400 pulses of stimulation. Data collected  
54  
55 from 1-4 seconds of signal registration were selected for integration, corresponding to a stimulation time  
56  
57 interval of 0.32 seconds, a value very similar to that used in CW-OSL (0.308s). The dose response curves for  
58  
59 TR-OSL (**Figure 2c**) follow the same pattern as those for CW-OSL, showing beyond reasonable doubt that the  
60  
61 difference observed between coarse and fine grains is inherent in quartz OSL, and not due to some contaminant.  
62  
63 The lifetimes were calculated for both natural and regenerated TR-OSL signals, as well as for signals obtained  
64  
65 after IR exposure and for the recycling point in the dose response curve, and these are shown as a function of  
66  
67 dose in **Figure 2d**. As in the previous study on ORL4 (Timar-Gabor et al., in press QI), the lifetimes for a

stimulation temperature of 125 °C are the same for both grain sizes and agree with the values of 36.5-38.5µs obtained by others (e.g. Chithambo, 2003; Ankjærgaard et al., 2010-figure 1) but there is no marked dependence of lifetime as function of given dose (c.f. Chithambo et al., 2008). These results indicate that the same recombination pathways are used to produce OSL in both fine and coarse grained quartz and these pathways are independent of dose. They also suggest that the second saturation exponential term observed in the dose response curve is not an effect of the existence of multiple recombination centres emitting within our detection window.

#### 4. Natural dose response

Following the approach of Chapot et al. (2012), we have calculated the average sensitivity-corrected natural signals down the section at Costineşti and plotted them as a function of the expected equivalent dose. The latter were calculated using the measured dose rate and the expected ages derived from a model based on changes in magnetic susceptibility down section (Constantin et al., 2014). A relative error of 10% was assigned to the paleomagnetic ages. Fine and coarse grains from 25 samples of loess from Costineşti with expected doses up to 500 Gy were used (Timar-Gabor and Wintle, 2013). These results are shown in **Figures 3a and 3b**, together with the value for “an infinitely old sample” (Timar-Gabor et al., 2012), with expected doses of between 2000 and 2500 Gy, depending on grain size. The natural dose response of the infinitely old sample is about 60% of the laboratory saturation level for fine grains and 80% of the laboratory saturation level in the case of coarse grains. These results provide further evidence that natural signals from infinitely old samples from this region do not fall on the laboratory dose response curves. By comparing **Figures 3a and 3b**, it is seen that the differences between the natural and laboratory dose response curves are more pronounced in the case of fine grains. Based on the dose response curves presented, the natural response seems to overestimate the laboratory response in the 100-400 Gy region, while for higher doses, as in the case of fine grains, the natural signals show earlier saturation.

#### 5. Other dose response curves

##### 5.1. Multiple-aliquot additive-dose with 110 °C TL correction

A new multiple-aliquot additive-dose (MAAD) procedure was applied, in which the OSL responses of all aliquots were normalised using the 110 °C TL peak measured in response to a test dose given before any other laboratory treatment, such as irradiation or preheating. Murray and Wintle (2000) pointed out that a subsequent test dose (as used in the standard for of SAR) did not monitor sensitivity change occurring during measurement of the natural OSL signal; they further showed that a prior 110°C TL measurement could be used for this purpose, if the changes in the sensitivity of the two signals were directly proportional, and Singhvi et al. (2011) later tested this suggestion. In our MAAD procedure, we used 96 aliquots of fine grains from CST 3, which had an equivalent dose of  $105 \pm 3$  Gy as derived using the standard SAR protocol. Immediately following delivery



of a 4 Gy test dose, the 110 °C TL peak was measured by heating to 180 °C. Subsequently the aliquots were divided into 12 groups of 8 aliquots and irradiated with doses ranging from 10 to 2580 Gy. Following a preheat for 10 s at 220 °C, the OSL was measured at 125 °C for 40 s, and this signal was normalised using the previously measured 110 °C TL response.

The MAAD dose response curve is given in **Figure 4** and the equivalent dose ( $103 \pm 20$  Gy) obtained by extrapolation of a single saturating exponential function (which was found to be the best fit for the presented dataset) is indistinguishable from the SAR result ( $105 \pm 3$  Gy). The scatter observed for each dose point is intriguing, possibly suggesting that individual aliquots have either a range of TL responses to the initial 4 Gy dose or different OSL responses to added dose. Either explanation is surprising, because fine grains are expected to have a homogenous response because of the large number of grains on each disc. Due to this high degree of scatter, a quantitative assessment based on fitting parameters of saturation characteristics cannot be performed with confidence. However, it is important to note that the MAAD dose response is not fully saturated for doses over 2000 Gy and thus grows to higher doses than the natural dose response, which is fully saturated at doses about 500-1000 Gy (**Figure 3**).

## 5.2. The effect of adding a large dose on top of natural signals

In an earlier study, Timar-Gabor and Wintle (2013) added 1816 Gy on top of the natural dose for fine grains from sample CST 6 ( $D_e = 197 \pm 7$  Gy) and 1676 Gy for coarse grains from another sample, CST 1 ( $D_e = 116 \pm 8$  Gy). The light levels obtained were compared with those reached by the laboratory dose response curve constructed using the SAR procedure using doses up to 2300 Gy. For the coarse grains, the signal seemed to be in saturation, while for fine grains the signal appeared to be below the maximum value obtained in the laboratory. However, the dose response curves did not show complete saturation. In our study, we have given additional doses of 8000 Gy to the fine grains and 5000 Gy to the coarse grains for the same sample, MST 7. Laboratory dose response curves were then constructed using doses up to 10000 Gy. For both grain sizes, signals from aliquots given added doses are below the saturation levels of the laboratory dose responses; this difference is greatest for fine grains (**Figure 5**). It seems likely that quartz cannot be used to accurately measure a known given dose at high doses (see **Figure S6** and **Table S3** -fine grains and **Figure S7** and **Table S4** -coarse grains for further information), and that signal instability is not likely to be the cause of underestimation of the natural signal relative to the saturation level in the infinitely old sample. In contrast, these results strongly suggest that, at large doses, sensitivity changes in the first measurement cycle are not adequately corrected for using the response to a test dose given either before or after measurement of the signal of interest. They also demonstrate the need to construct laboratory dose response curves up to full saturation before pertinent conclusions regarding the saturation parameters can be drawn.

## 6. Summary and Conclusions

1 SAR measurement sequences carried out on fine (4-11  $\mu\text{m}$ ) and coarse (63-90  $\mu\text{m}$ ) grained quartz extracts from  
 2  
 3 loess deposits at four sites in the south of Romania have confirmed previously published work that shows the  
 4  
 5 SAR laboratory dose response curves obtained for the two grain sizes to be quite different. For both grain sizes,  
 6  
 7 the sum of two exponential functions is required to fit the data, characterised by  $D_{01}$  and  $D_{02}$  values of  $\sim 140$  and  
 8  
 9  $\sim 1400$  Gy for fine grains and  $\sim 65$  and  $\sim 650$  Gy in the case of coarse grains, respectively, when doses up to 5000  
 10 Gy are given (**Figure 1**). Only when the laboratory dose was extended up to 9 kGy is the fine-grain signal seen  
 11 to be in full saturation. Laboratory saturation characteristics are not affected by previous irradiation, bleaching,  
 12  
 13 or the thermal treatments employed.  
 14  
 15 TR-OSL measurements on fine and coarse grains showed signals typical of quartz (**Figures 2a and b**), with  
 16  
 17 lifetime values independent of dose (**Figure 2d**). These results imply that the second exponential component in  
 18  
 19 the dose response curve is not a result of a varying contribution from different luminescent centres.  
 20  
 21 For doses higher than about 200 Gy natural and laboratory dose response curves for fine grains diverge (**Figure**  
 22  
 23 **3a**), with natural signals showing earlier saturation. Based on the divergence shown for these samples from  
 24  
 25 Costinești, it is tempting to suggest that an upper limit of  $2 D_{01}$  could be applied to values of  $D_e$  determined  
 26  
 27 using SAR, as suggested by Wintle and Murray (2006) for single exponential growth. For these samples, Timar-  
 28  
 29 Gabor and Wintle (2013, Figure 2c) obtained a value of  $D_{01} = 76$  Gy when they fitted a double exponential to  
 30  
 31 the average dose response curve for all samples from this section constructed using doses up to 1200 Gy.  
 32  
 33 However, as shown in **Figure 1a**,  $D_{01}$  ranges between  $131 \pm 20$  and  $200 \pm 24$  Gy when doses up to 5000 Gy  
 34  
 35 were used to construct the dose response curve; for  $\text{CST}_{\text{inf}}$ , the value was  $144 \pm 19$  Gy. It is thus not possible to  
 36  
 37 provide firm numerical guidance on the maximum value of  $D_e$  that can be determined reliably. In addition, the  
 38  
 39 comparison is constrained by the uncertainty in the  $D_e$  predicted down section based on the dose rate and age  
 40  
 41 from the palaeomagnetic model, though likely error bars are given in **Figure 1a**. For 63-90  $\mu\text{m}$  quartz, it is  
 42  
 43 difficult to assess the degree of overlap of the natural and laboratory dose response due to the scatter of the  
 44  
 45 datapoints (**Figure 3b**).  
 46  
 47 The multiple-aliquot additive dose response curve (**Figure 4**) seems to grow to higher doses than the natural  
 48  
 49 dose response; while there is agreement between the values obtained by SAR and MAAD for equivalent doses  
 50  
 51  $\sim 100$  Gy, the signals measured after adding a dose of the magnitude of the SAR saturation level (5000 Gy for  
 52  
 53 coarse grains; 8000 Gy for fine grains) on top of the natural dose were found to be significantly below the  
 54  
 55 saturation level (**Figure 5**). This suggests an initial dose-dependent sensitivity change that is not corrected for  
 56  
 57 by the use of the response to a test dose, whether given before or after the signal of interest.  
 58  
 59 In conclusion, several lines of investigation, such as signal contamination and the effects of prior dose and  
 60  
 61 thermal treatments, were unable to identify the cause of either the differences between quartz fine-grained and  
 62  
 63 coarse-grained dose response curves, or the age under-estimation at high doses. Although our previous studies  
 64  
 65 have shown that acceptable recovered-to-given dose ratios could be obtained up to  $\sim 500$  Gy (Timar-Gabor et  
 al., 2011, Timar-Gabor et al., 2012, supplementary material) or even  $\sim 1500$  Gy (Constantin et al., 2014) when

doses are given after a laboratory bleach, our present study suggests that age underestimation may, in part, be caused by poor dose recovery at very high doses. Further investigations in this high dose range are clearly needed and it is recommended that adding a large dose on top of the natural and comparing the measured signal to the laboratory saturation limit should be incorporated as a standard test for evaluating the performance of the SAR protocol, especially when applications are carried out in the high dose range.

## Acknowledgements

A.T-G. and D.C. acknowledge the financial support from a grant of the Romanian National Authority for Scientific Research CNCS-UEFISCDI, PN-II-RU-TE-2011-3-0062, nr. 73/05.10.2011.D.C. also acknowledges the financial support of the Sectorial Operational Programme for Human Resources Development 2007-2013, co-financed by the European Social Fund under the project POSDRU/159/1.5/S/133391-“Doctoral and postdoctoral excellence programs for training highly qualified human resources for research in the fields of Life Sciences, Environment and Earth”.

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### **Figure captions**

**Figure 1:** Laboratory dose response of (a) 15 aliquots of fine grained (4-11  $\mu\text{m}$ ) quartz and (b) 11 aliquots of coarse grained (63-90  $\mu\text{m}$ ) quartz. For fine grains, the individual values for  $D_{01}$  range between  $131 \pm 20$  Gy and  $200 \pm 24$  Gy, while the  $D_{02}$  values are between  $1252 \pm 119$  Gy and  $1917 \pm 280$  Gy. In the case of coarse grain, values for  $D_{01}$  range between  $30 \pm 3$  Gy and  $68 \pm 6$  Gy while the minimum and maximum values for  $D_{02}$  are  $317 \pm 26$  Gy and  $705 \pm 120$  Gy. The average dose response curve for each grain size is represented as a solid line.

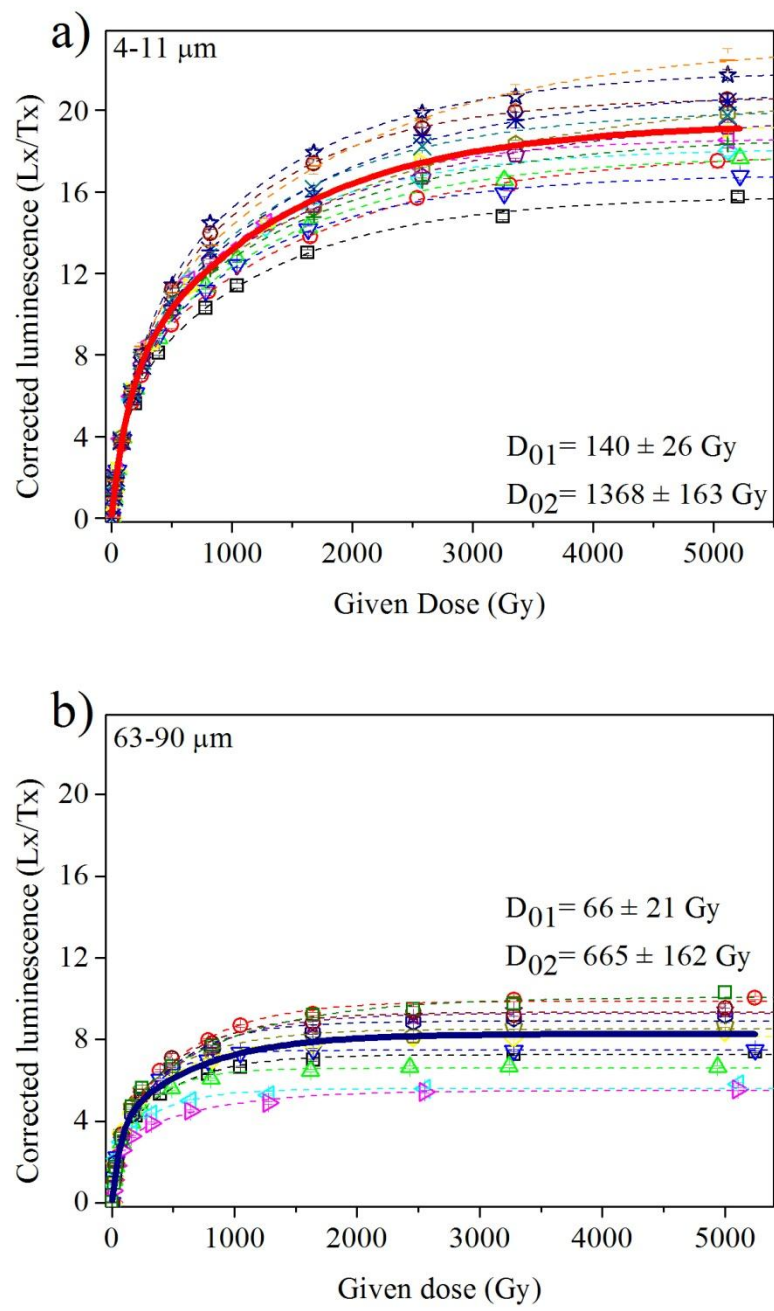
**Figure 2:** TR-OSL signals (photon arrival time distributions-PAT) for one representative aliquot of (a) fine and (b) coarse grains of sample CST 3 recorded after different doses. (c) Dose response curves for fine (4-11  $\mu\text{m}$ ) grains and coarse (63-90  $\mu\text{m}$ ) grains of sample CST 3 constructed using TR-OSL. (d) Average lifetimes obtained as function of dose. The lifetimes of natural signals are depicted as stars. The IR stimulation was performed in a pseudo-CW stimulation manner (on time 500  $\mu\text{s}$ ; off time 4.5  $\mu\text{s}$ ) for 100 s at 125 °C. Each datapoint represents the average value obtained on 10 aliquots.

1 **Figure 3:** The average sensitivity-corrected natural signal for 26 samples from Costinești **(a)** fine and **(b)** coarse  
2 grains plotted as function of their expected equivalent dose. The average laboratory dose response curve (based  
3 on data taken up to 5000 Gy) is displayed for each grain size.  
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8 **Figure 4:** Multiple aliquot additive dose response curve obtained using 110 °C TL peak for normalisation.  
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12 **Figure 5:** Normalised luminescence signals for sample MST 7 ( $De = 149 \pm 3$  Gy- fine grains;  $De = 191 \pm 17$   
13 Gy- coarse grains); following irradiation to a dose of **(a)** 8000 Gy for fine grains and **(b)** 5120 Gy for coarse  
14 grains given on top of the natural dose.  
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Figure 1



Figure

Figure 2

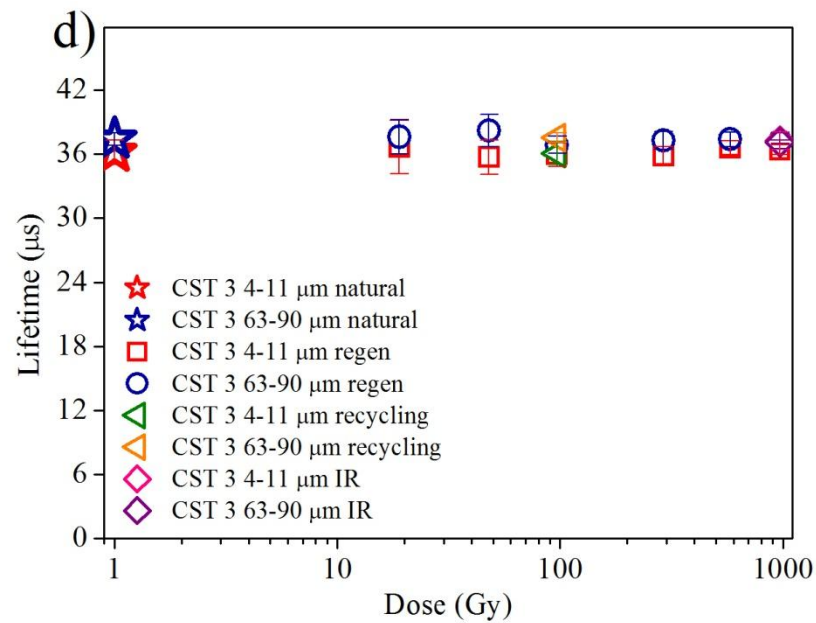
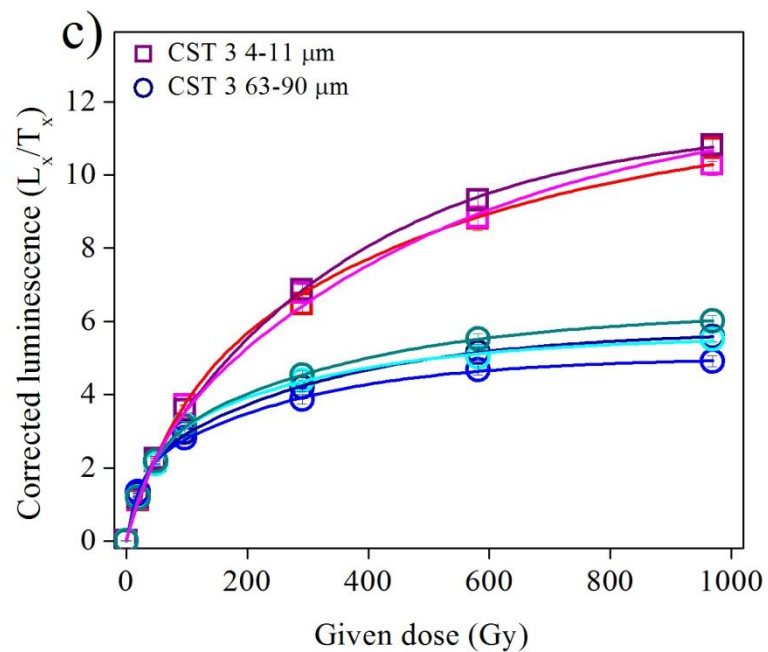
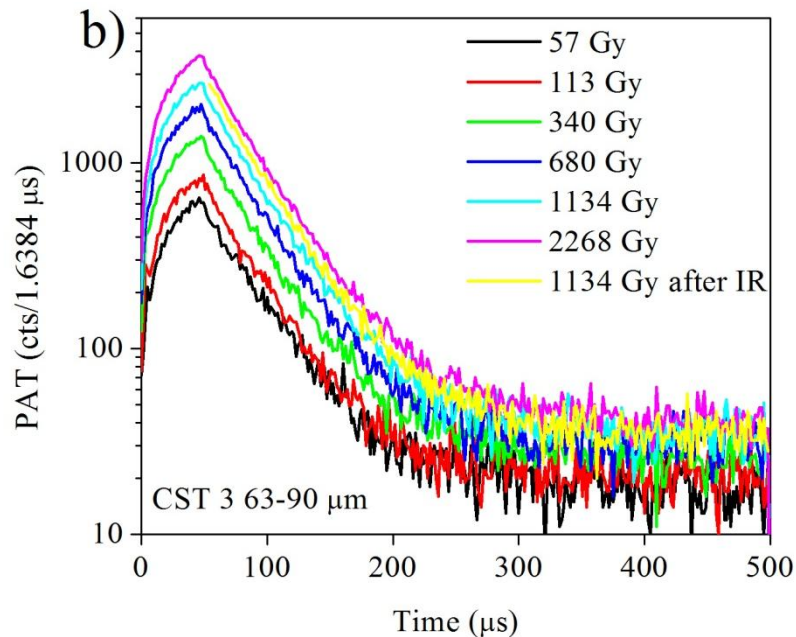
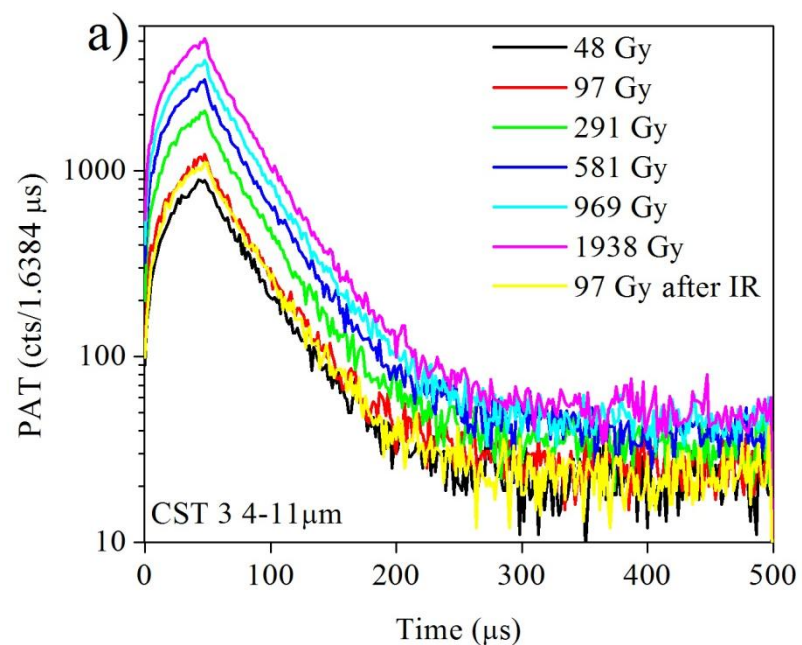




Figure 3

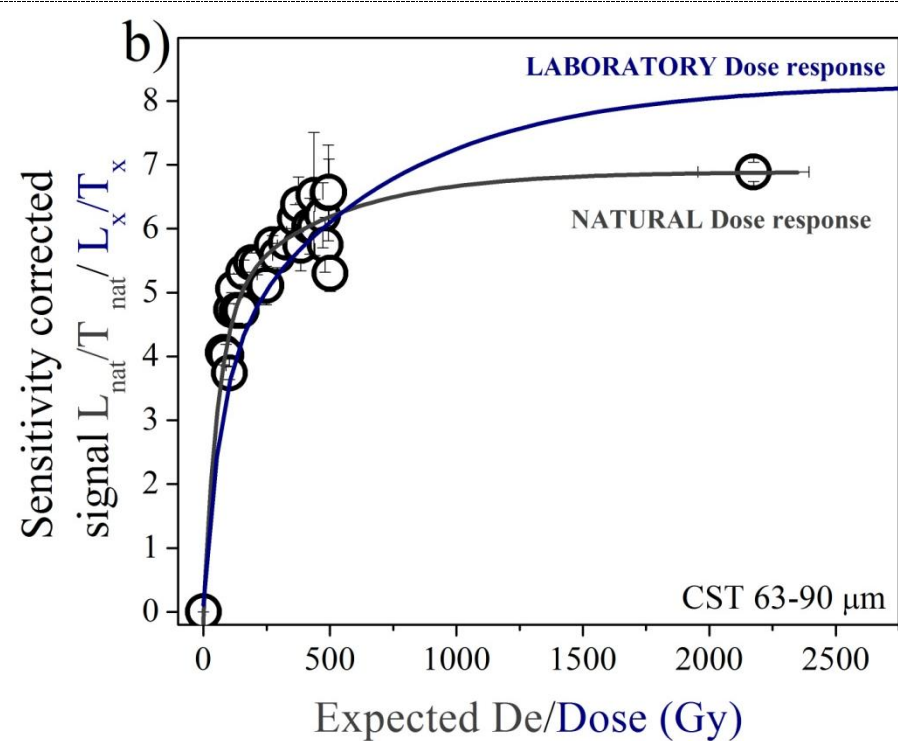
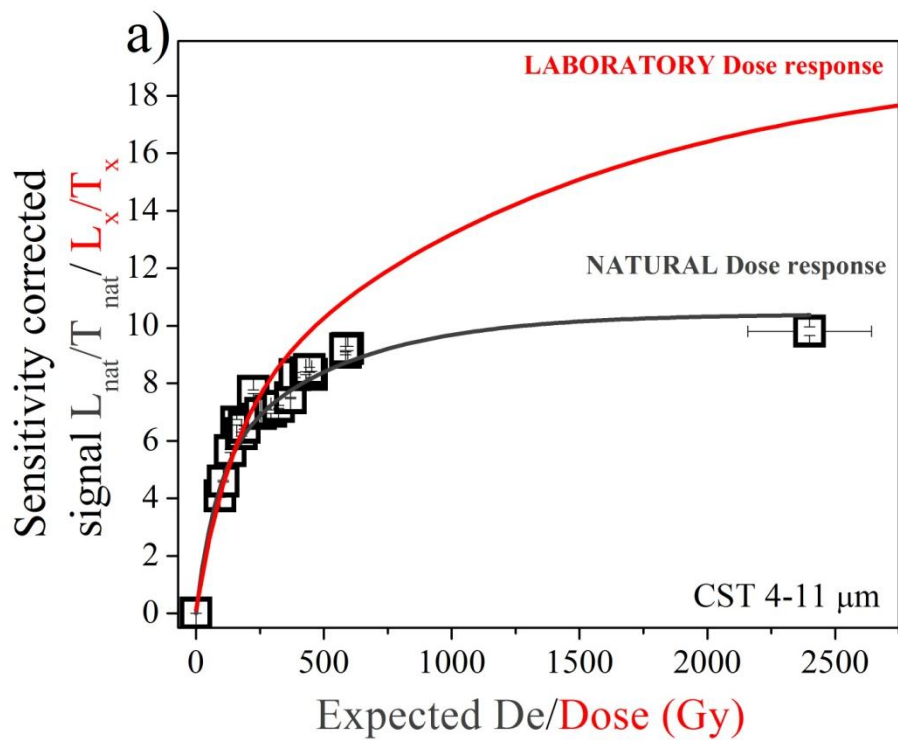
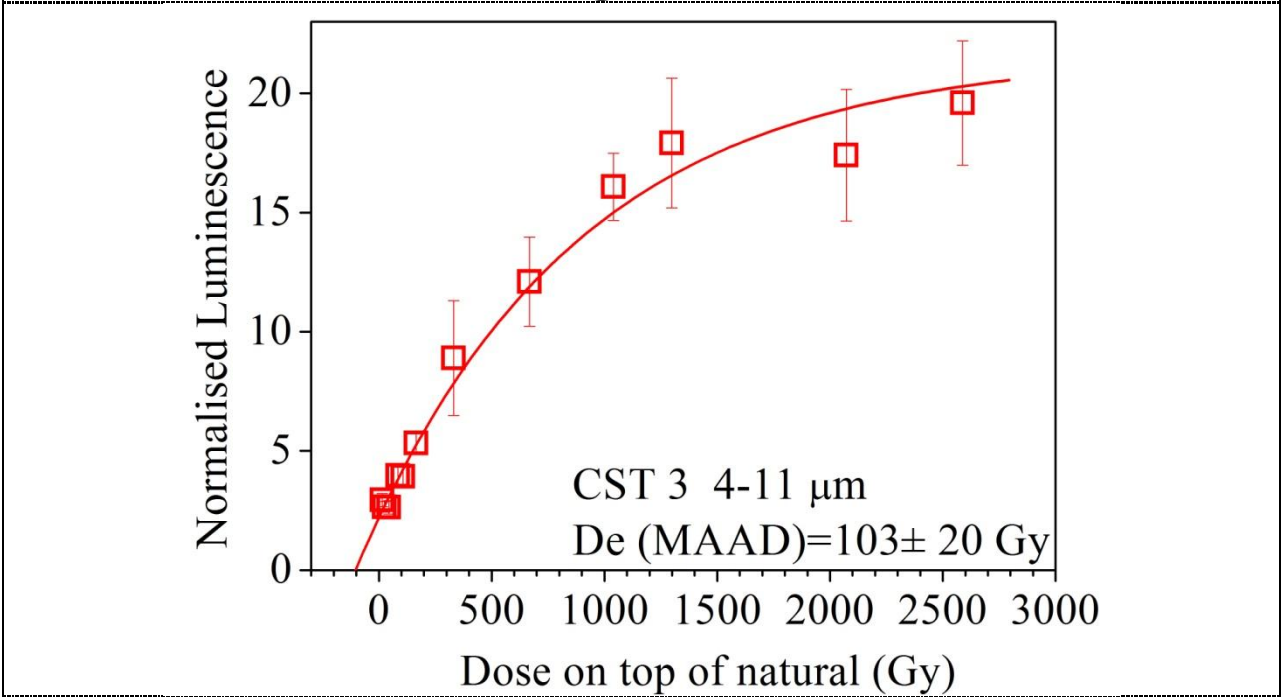
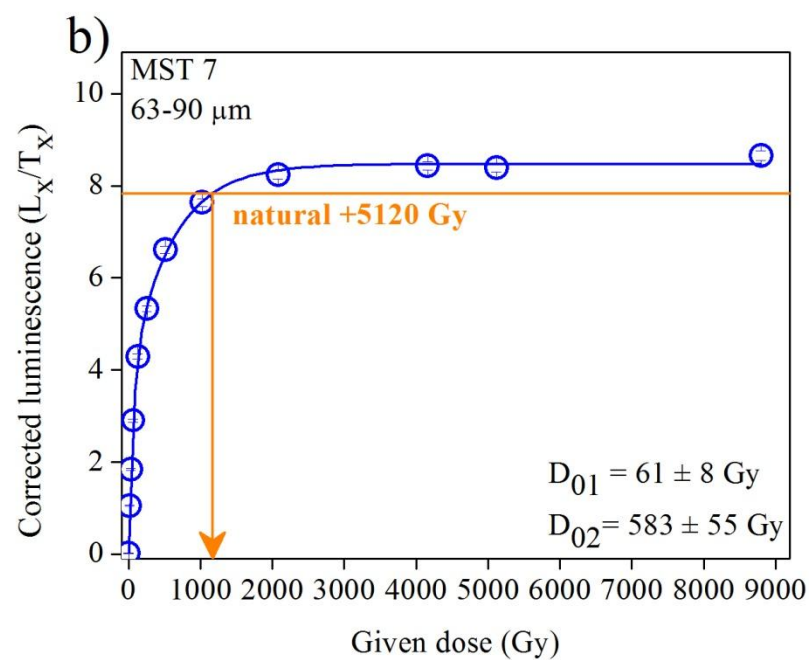
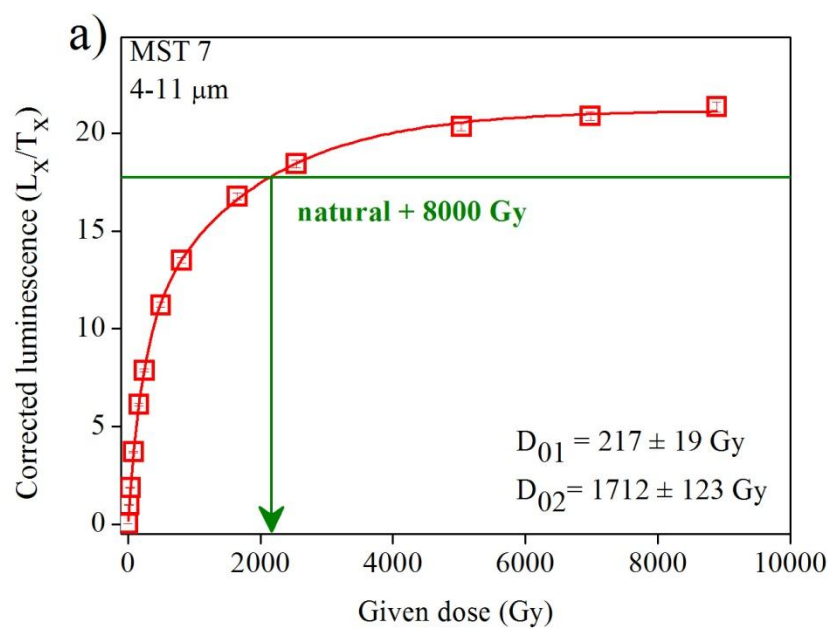


Figure 4



Figure

Figure 5

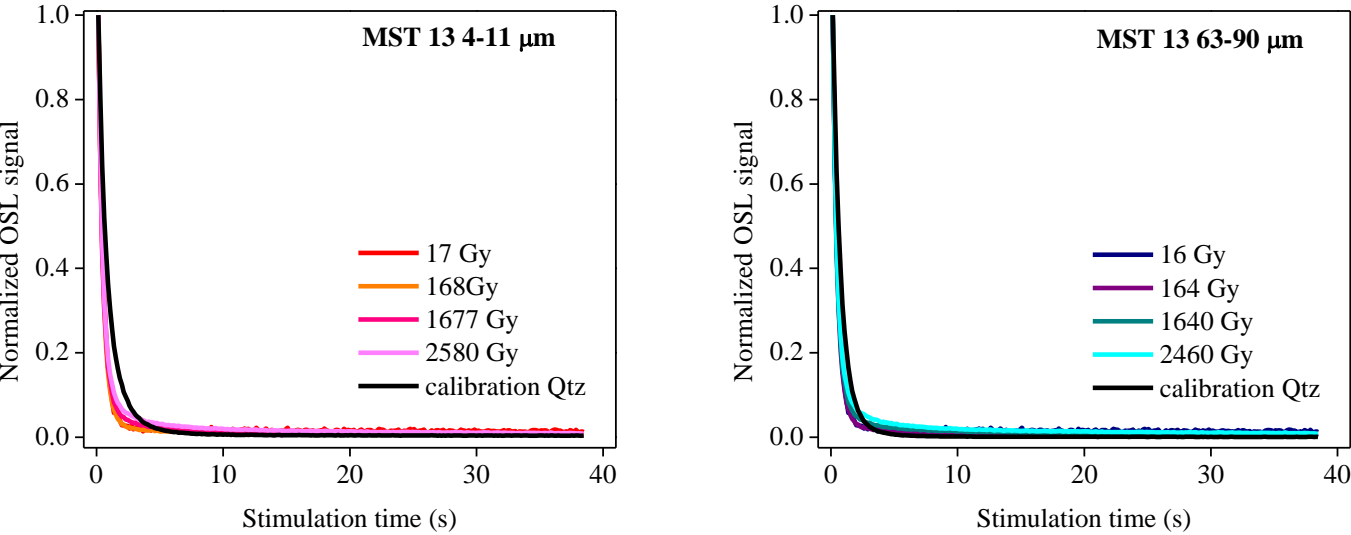


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**SUPPLEMENTARY- Fundamental investigations of natural and laboratory generated SAR dose response curves for quartz OSL in the high dose range**

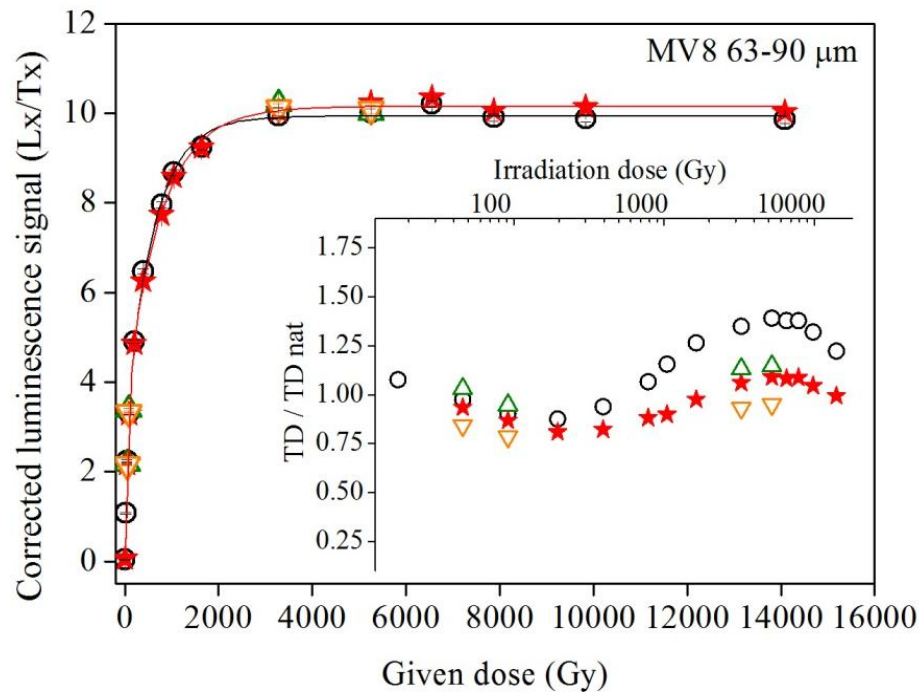
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**Table S1:** Additional information on investigated samples:

Sample code	De 4-11 $\mu\text{m}$ quartz (Gy)	De 63-90 $\mu\text{m}$ quartz (Gy)	Reference
MV 2	$32 \pm 1$	$46 \pm 4$	Timar-Gabor et al. 2011
MV 8	$209 \pm 3$	$247 \pm 8$	Timar-Gabor et al. 2011
MV 10	$310 \pm 9$	$319 \pm 15$	Timar-Gabor et al. 2011
CAC 2.1.	$126 \pm 4$	$119 \pm 9$	Constantin et al. 2012
MST 2	$113 \pm 2$	$130 \pm 10$	Vasiliniuc et al., 2011; Timar-Gabor et al. 2012
MST 3	$151 \pm 4$	$153 \pm 4$	Vasiliniuc et al., 2011; Timar-Gabor et al. 2012
MST 6	$147 \pm 2$	$185 \pm 9$	Vasiliniuc et al., 2011; Timar-Gabor et al. 2012
MST 7	$149 \pm 3$	$191 \pm 17$	Vasiliniuc et al., 2011; Timar-Gabor et al. 2012
MST 13	$195 \pm 2$	$277 \pm 15$	Vasiliniuc et al., 2011; Timar-Gabor et al. 2012
MST 17	$264 \pm 9$	$279 \pm 28$	Vasiliniuc et al., 2011; Timar-Gabor et al. 2012
MST 19	$289 \pm 9$	$288 \pm 57$	Vasiliniuc et al., 2011; Timar-Gabor et al. 2012
LCA 15	$113 \pm 2$	$117 \pm 5$	Constantin et al. in press
CST 1 $\rightarrow$ CST 25	$90 \pm 1 \rightarrow 409 \pm 12$	$116 \pm 8 \rightarrow 470 \pm 38$	Constantin et al. 2014, Timar-Gabor and Wintle 2013
Cst inf.	$650 \pm 20$	$628 \pm 47$	Timar-Gabor et al. 2012

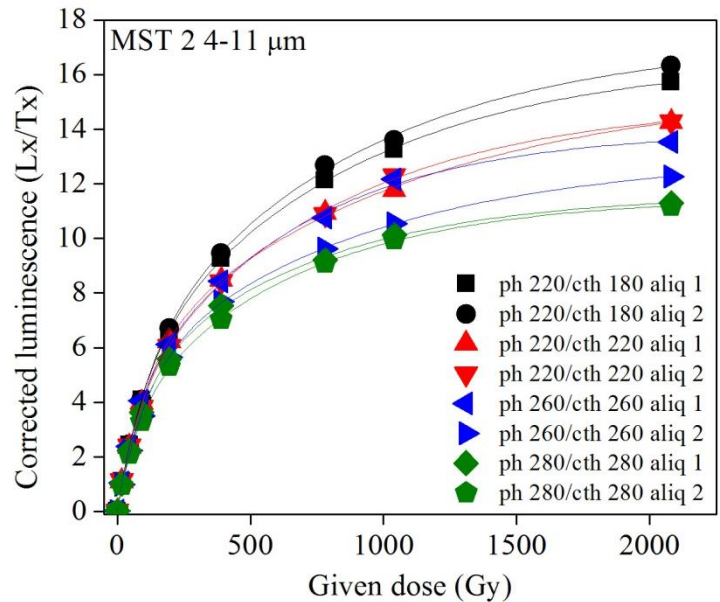
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**Figure S1:** CW-OSL decay curves as a function of dose compared to the calibration quartz (4.81 Gy) OSL decay curve. The data are normalized to the number of counts in the first channel (first 0.15 s of the stimulation time).



**Figure S2:** Dose response curve reproducibility test for a coarse quartz aliquot of sample MV8 (see text for experimental details). The first growth is represented by circles while the second growth is represented by stars. Repeat points for the first and second dose responses are represented by up and down triangles respectively. The inset shows the change in sensitivity monitored by the response to the test dose.



**Figure S3:** Dose response curves constructed up to 2000 Gy using different combinations of preheat/cutheat treatments, for a sample of fine quartz. Fitting parameters in the case of this particular sample, as well as the coarse quartz counterpart material, are given in the table below.



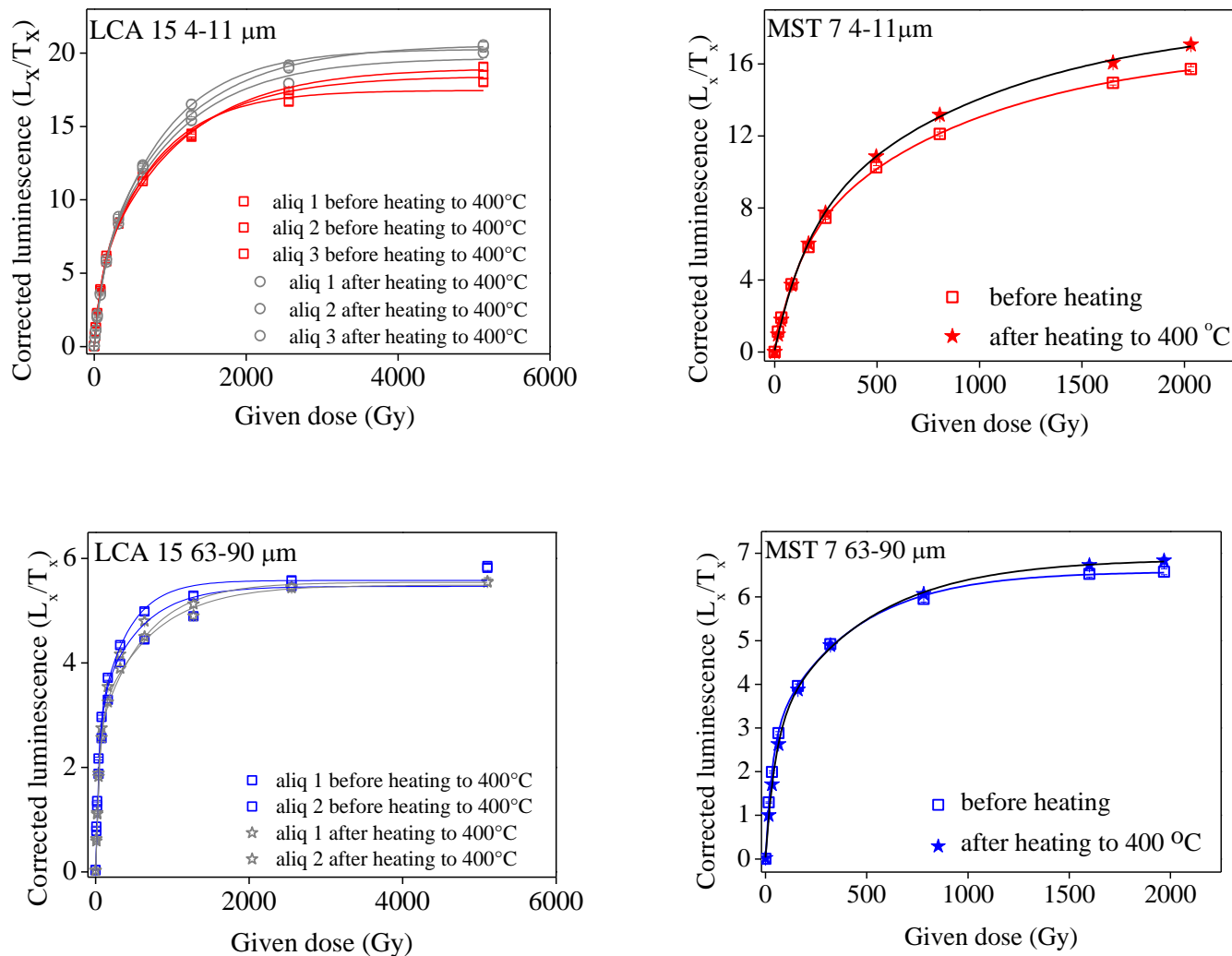
# SUPPLEMENTARY- Fundamental investigations of natural and laboratory generated SAR dose response curves for quartz OSL in the high dose range

**Table S2:** Fitting parameters for OSL dose response curves constructed up to 2000 Gy for different thermal treatments. Please note that fitting parameters should be regarded as an indication of a specific trend rather than a quantitative measure, due to the restricted number of points used and dose range investigated.

Sample	$y_0$	$y_0$ error	$A_1$	$A_1$ error	$D_{01}$	$D_{01}$ error	$A_2$	$A_2$ error	$D_{02}$	$D_{02}$ error	Reduced $\chi^2$	$R^2$
<b>4 – 11 <math>\mu</math>mMST 2</b>												
<b>Ph 220 cth 180</b>	0.09	0.07	4.8	0.4	<b>105</b>	11	11.7	0.3	<b>826</b>	56	0.007	1.000
<b>Ph 220 cth 180</b>	0.08	0.12	5.0	0.8	<b>112</b>	20	12.2	0.6	<b>836</b>	101	0.020	0.999
<b>Ph 220 cth 220</b>	0.09	0.09	5.3	0.5	<b>108</b>	13	10.2	0.3	<b>1035</b>	131	0.012	1.000
<b>Ph 220 cth 220</b>	0.08	0.11	3.8	0.5	<b>82</b>	14	11.1	0.4	<b>761</b>	68	0.016	0.999
<b>Ph 260 cth 260</b>	0.02	0.13	3.6	0.5	<b>72</b>	15	10.3	0.4	<b>629</b>	59	0.021	0.999
<b>Ph 260 cth 260</b>	0.08	0.07	4.7	0.4	<b>107</b>	12	8.2	0.3	<b>871</b>	94	0.007	1.000
<b>Ph 280 cth 280</b>	0.07	0.08	4.2	0.4	<b>91</b>	11	7.4	0.3	<b>675</b>	64	0.008	1.000
<b>Ph 280 cth 280</b>	0.07	0.07	3.4	0.3	<b>82</b>	10	8.1	0.3	<b>645</b>	45	0.006	1.000
Sample	$y_0$	$y_0$ error	$A_1$	$A_1$ error	$D_{01}$	$D_{01}$ error	$A_2$	$A_2$ error	$D_{02}$	$D_{02}$ error	Reduced $\chi^2$	$R^2$
<b>63-90 <math>\mu</math>mMST 3</b>												
<b>Ph 220 cth 180</b>	0.09	0.07	3.2	0.3	<b>47</b>	8	3.9	0.3	<b>762</b>	163	0.02	1.00
<b>Ph 220 cth 180</b>	0.08	0.12	2.4	0.6	<b>41</b>	17	4.2	0.5	<b>464</b>	122	0.05	0.99
<b>Ph 220 cth 220</b>	0.09	0.09	1.7	0.3	<b>28</b>	8	5.3	0.3	<b>252</b>	22	0.01	1.00
<b>Ph 220 cth 220</b>	0.08	0.11	3.0	0.4	<b>50</b>	11	3.7	0.3	<b>657</b>	157	0.02	1.00
<b>Ph 260 cth 260</b>	0.02	0.13	2.2	0.2	<b>28</b>	4	2.2	0.2	<b>256</b>	36	0.00	1.00
<b>Ph 260 cth 260</b>	0.08	0.07	2.1	0.3	<b>38</b>	10	5.8	0.3	<b>431</b>	45	0.01	1.00
<b>Ph 280 cth 280</b>	0.07	0.08	1.7	1.0	<b>29</b>	17	2.0	1.0	<b>145</b>	68	0.02	0.99
<b>Ph 280 cth 280</b>	0.07	0.07	2.8	0.4	<b>45</b>	9	1.5	0.3	<b>394</b>	162	0.01	0.99

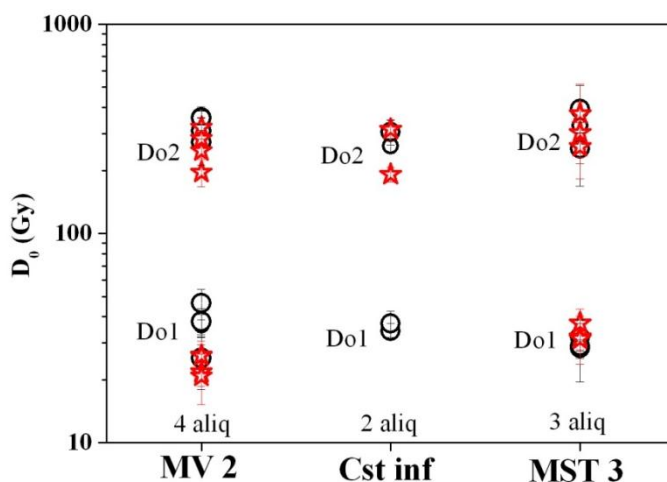
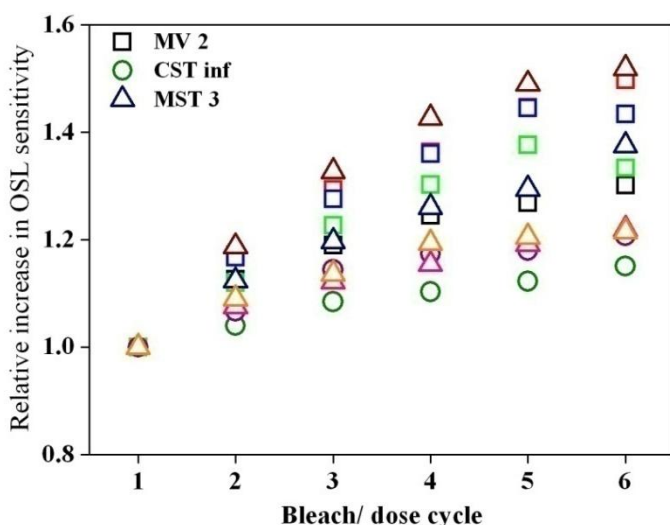
# SUPPLEMENTARY- Fundamental investigations of natural and laboratory generated SAR dose response curves for quartz OSL in the high dose range

**Figure S4:** Dose response curves for fine and coarse quartz grains constructed before and after heating the sample to 400 °C.



# SUPPLEMENTARY- Fundamental investigations of natural and laboratory generated SAR dose response curves for quartz OSL in the high dose range

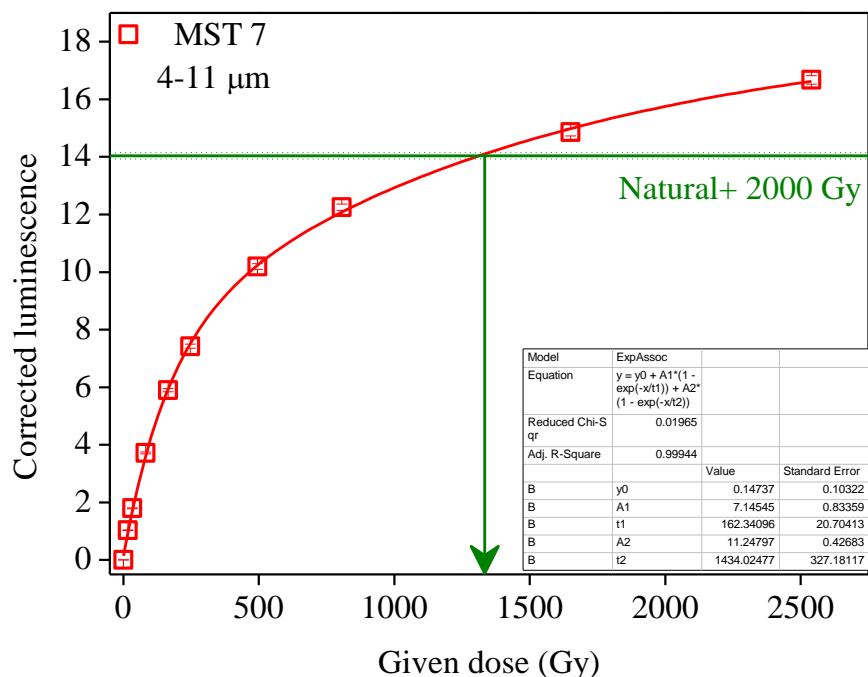
**Figure S5:** (a) Variations in OSL sensitivity as a function of bleach/dose cycle obtained using nine coarse quartz aliquots of samples collected from Mircea Voda, Costinesti and Mostistea. The samples were first optically zeroed using a double blue stimulation at room temperature for 100 s with 10 ks pause in between. The aliquots were then given a 170 Gy dose and the OSL signal was measured using a blue stimulation at 125 °C for 40s. This was repeated for five times in order to accumulate a dose of ~ 1000 Gy after bleaching. A dose-response curve was then built using seven regenerative doses up to ~ 1050 Gy. The data represent the net OSL signals obtained in each cycle normalized to the OSL signal from the first cycle. (b) Comparison between the saturation characteristics ( $D_{01}$ ,  $D_{02}$ ) obtained for dose-response curves constructed after multiple bleach/dose cycles (open stars) and the values obtained for dose-response curves constructed for  $D_e$  measurement, on different aliquots. It can be noted that there is a difference between the individual values (which is to be expected, due to the fact that different aliquots of coarse grains are investigated ; i.e. see scatter in fig 1.b), but remain consistent within error limits.





# SUPPLEMENTARY- Fundamental investigations of natural and laboratory generated SAR dose response curves for quartz OSL in the high dose range

**Figure S6:** Normalised luminescence signal for sample MST 7 4-11  $\mu\text{m}$  quartz, with an equivalent dose of 149 Gy following irradiation with a dose of 2000 Gy.



**Table S3:** Further results of adding a laboratory dose on top of natural doses for fine quartz grains of samples MST7, MST11 and MST 17.

## Sample MST 7 4-11 $\mu\text{m}$ $D_e=149\pm3$ Gy

Given dose (Gy)	MEASURED DOSE	Nr of aliq	Measured/Given dose
"NATURAL"	$149 \pm 3$ Gy	6	-
NAT + 99 Gy=248Gy	$236 \pm 3$ Gy	2	$0.95\pm0.02$
	$237 \pm 3$ Gy		
NAT + 198 Gy=347Gy	$348 \pm 5$ Gy	2	$0.99\pm0.02$
	$337 \pm 5$ Gy		
NAT + 397 Gy=546Gy	$491 \pm 8$ Gy	2	$0.89\pm0.02$
	$485 \pm 10$ Gy		
NAT + 595 Gy=744Gy	$699 \pm 16$ Gy	2	$0.95\pm0.02$
	$718 \pm 15$ Gy		
NAT + 977 Gy=1126 Gy	$1088 \pm 25$ Gy	3	$0.96\pm0.03$
	$1028 \pm 17$ Gy		
	$149 \pm 3$ Gy		
NAT + 2000 Gy=2149 Gy	$1373\pm24$ Gy	1	$0.64\pm0.01$
NAT + 4508 Gy=4657 Gy	$1989\pm50$ Gy	1	$0.43\pm0.01$
NAT + 7938 Gy=8087 Gy	$2930\pm83$ Gy	1	$0.36\pm0.01$

## Sample MST 11 $\mu\text{m}$ $D_e=173\pm4$ Gy

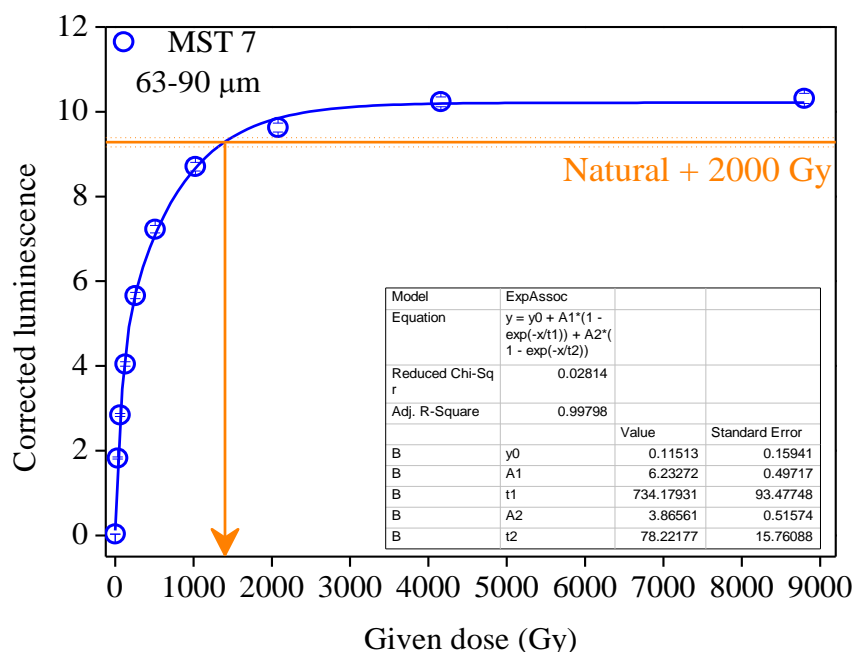
Given dose (Gy)	MEASURED DOSE	Nr of aliq	Measured/Given dose
"NATURAL"	173 Gy	10	-
NAT + 175 Gy=348 Gy	$341 \pm 11$ Gy	3	$0.98 \pm 0.04$

## Sample MST 17 $\mu\text{m}$ $D_e=264\pm9$ Gy

Given dose (Gy)	MEASURED DOSE	Nr of aliq	Measured/Given dose
"NATURAL"	$264 \pm 9$ Gy	7	-
NAT + 705 Gy=969 Gy	$924 \pm 51$ Gy	1	$0.95 \pm 0.06$

# SUPPLEMENTARY- Fundamental investigations of natural and laboratory generated SAR dose response curves for quartz OSL in the high dose range

**Figure S7:** Normalised luminescence signal for sample MST 7 63-90 µm quartz, with an equivalent dose of 170 Gy following irradiation with a dose of 2000 Gy.



**Table S4:** Further results of adding a laboratory dose on top of natural doses for coarse quartz grains of sample MST7.

**Sample MST 7 63-90 µm De=170± 8Gy n=9**

Given dose (Gy)	MEASURED DOSE	Nr of aliq	Measured/ Given dose
“NATURAL”	170 ± 8 Gy	8	-
NAT + 5000 Gy	1121 Gy	1	0.22
NAT + 2000 Gy	1197 Gy	1	0.55
NAT + 800 Gy	830 Gy	1	0.86
NAT +200 Gy	330 Gy	1	0.89
NAT +104 Gy	258 ± 12 Gy	5	0.94±0.06